Resonant x-ray diffraction in manganites: what can we learn about the coupling between magnetic and orbital order?

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ALS User Meeting 2005

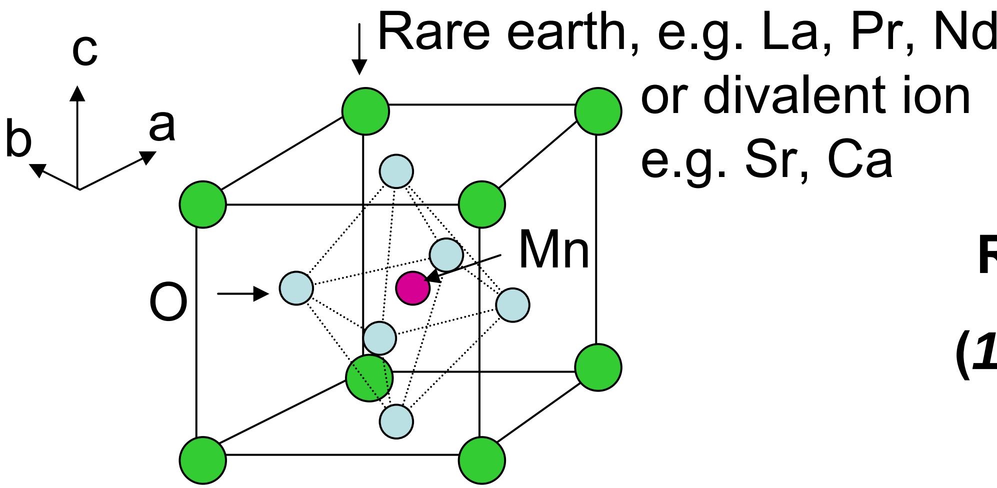
Overview

1. *Charge, orbital and spin ordering in half-doped manganites – open questions.*

2. *Comparison of orbital and magnetic correlations (soft x-ray resonant diffraction).*

3. *Coherent resonant x-ray diffraction: exploring orbital domain evolution and dynamics.*

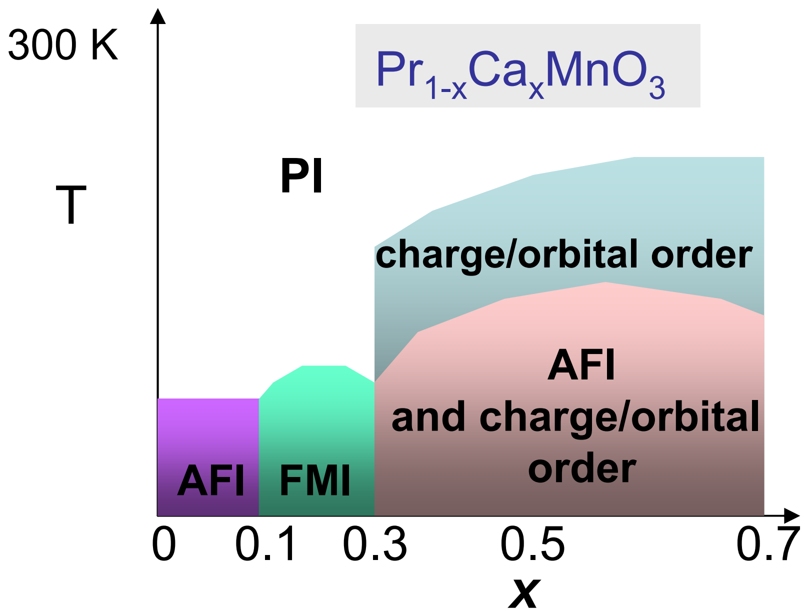
Manganites: structure and properties



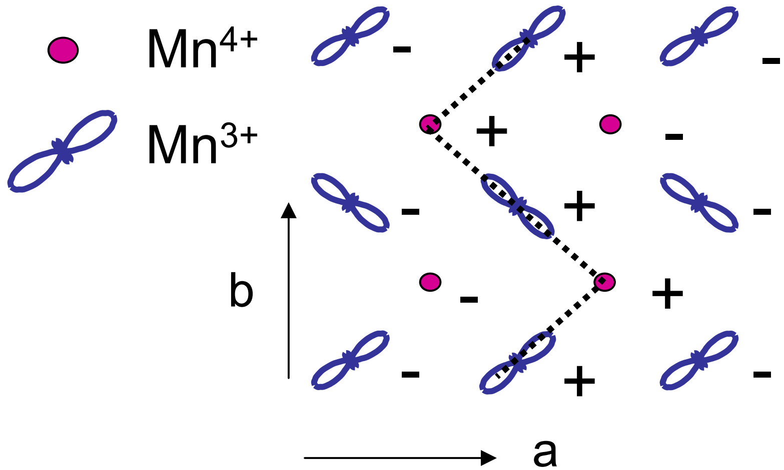
$$RE_{1-x}A_xMnO_3 \text{ (e.g. } La_{1-x}Sr_xMnO_3)$$

$$(1-x)RE^{3+} + xA^{2+} + Mn^{v} + 3(O^{2-}) = 0$$

$$v = 3 + x$$



Charge, orbital and magnetic order in half-doped manganites

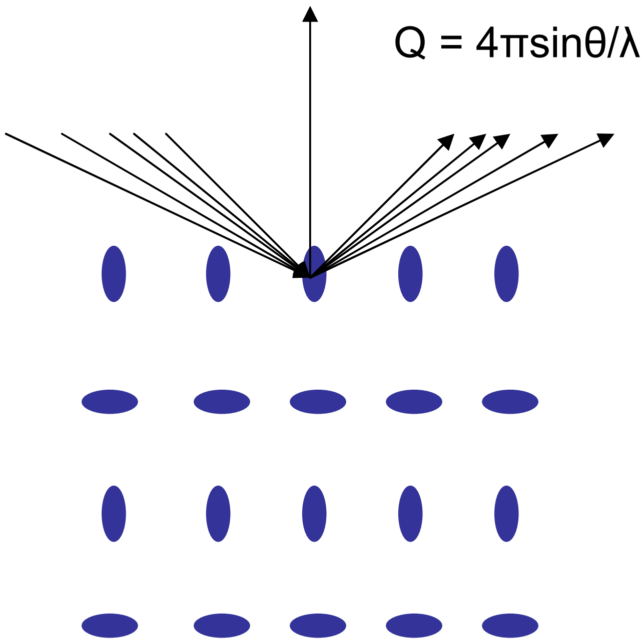


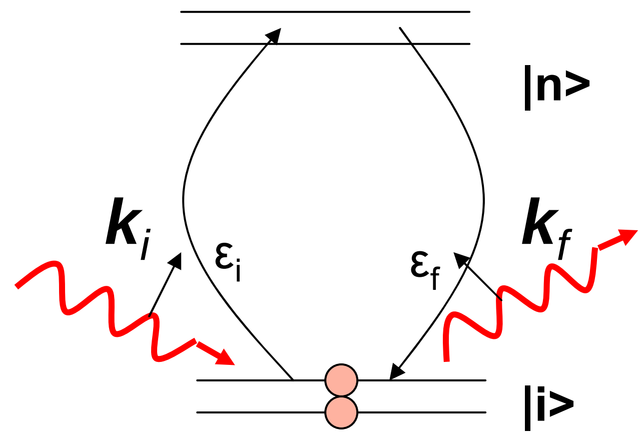
Goodenough (1955)

Fundamental questions

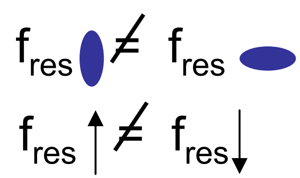
- **What drives charge/orbital order?**
- **Coupling between orbital and magnetic correlations?**
- **What sets the length scale for the correlation lengths?**

Resonant x-ray diffraction: a means for measuring subtle modulations





$$f_{\text{res ion}} \sim \frac{\sum_n \langle i\,|\boldsymbol{\varepsilon}|n\rangle\langle n|\boldsymbol{\varepsilon}|i\rangle}{E - E_{res} + i\Gamma/2}$$



$$f_{ion} = f(Q) + f'(E) + if''(E)$$

$$I = |\Sigma\, f_{ion} e^{(i\mathbf{Q}\cdot\mathbf{r})}|^2$$

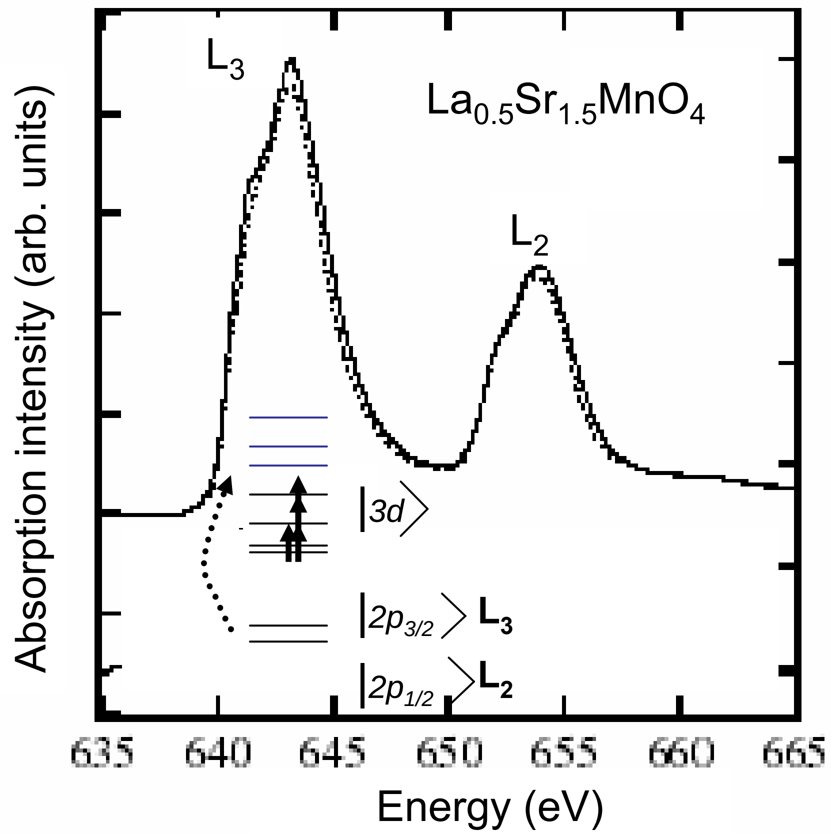
- Element specific
- Sensitive to atomic environment (lattice distortions, electron occupancy, spin)
- Enhances scattering (forbidden/nearly forbidden reflections, magnetic scattering)

Mn 2p -> 3d L-edge

*C. W. M. Castleton and M. Altarelli, PRB **62**, 1033 (2000).*

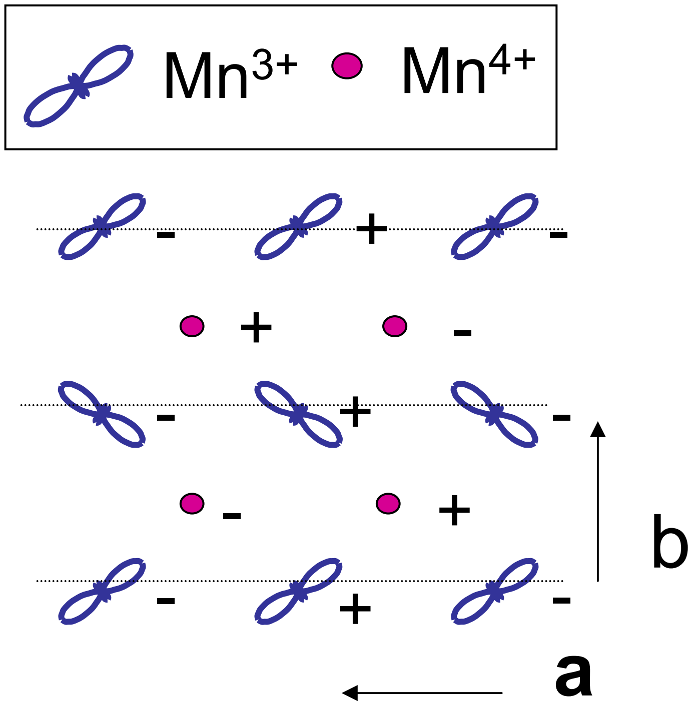
Using L-edge diffraction would provide information on the type of orbital ordering as well as permitting "the effects of orbital ordering and Jahn-Teller ordering to be detected and distinguished from one another."

E ~ 650 eV, λ ~ 19 Å

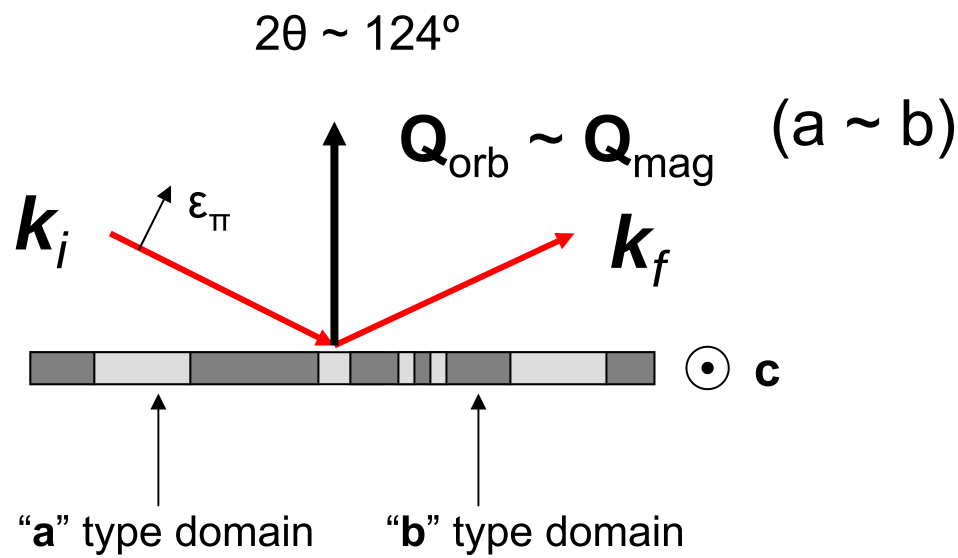


D. J. Huang et al., PRL 92 087202 (2004)

Crystal is twinned with both [100] and [010] surface normal domains.
→ Magnetic and orbital scattering observed at the same scattering angle



$\mathbf{Q}_{orb}$ = 2π/b (0,½,0)
$\mathbf{Q}_{mag}$ = 2π/a (½,0,0)

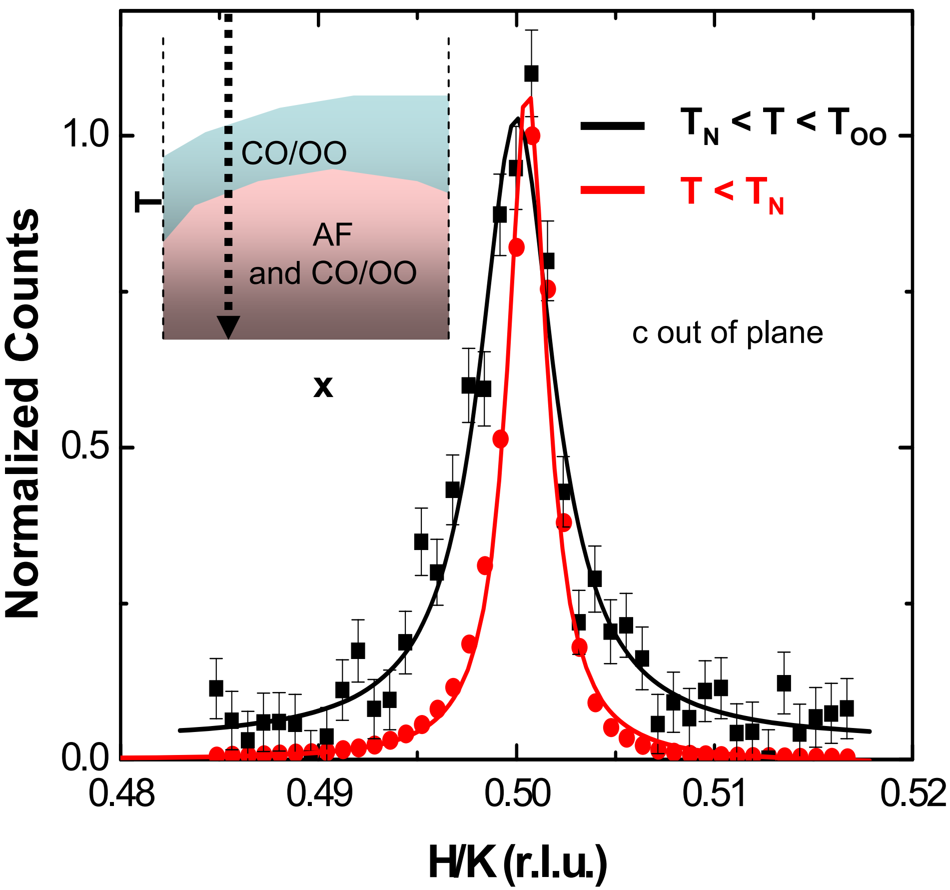


Magnetic and orbital scattering not resolved in Q

Direct comparison of orbital and magnetic superlattice reflections

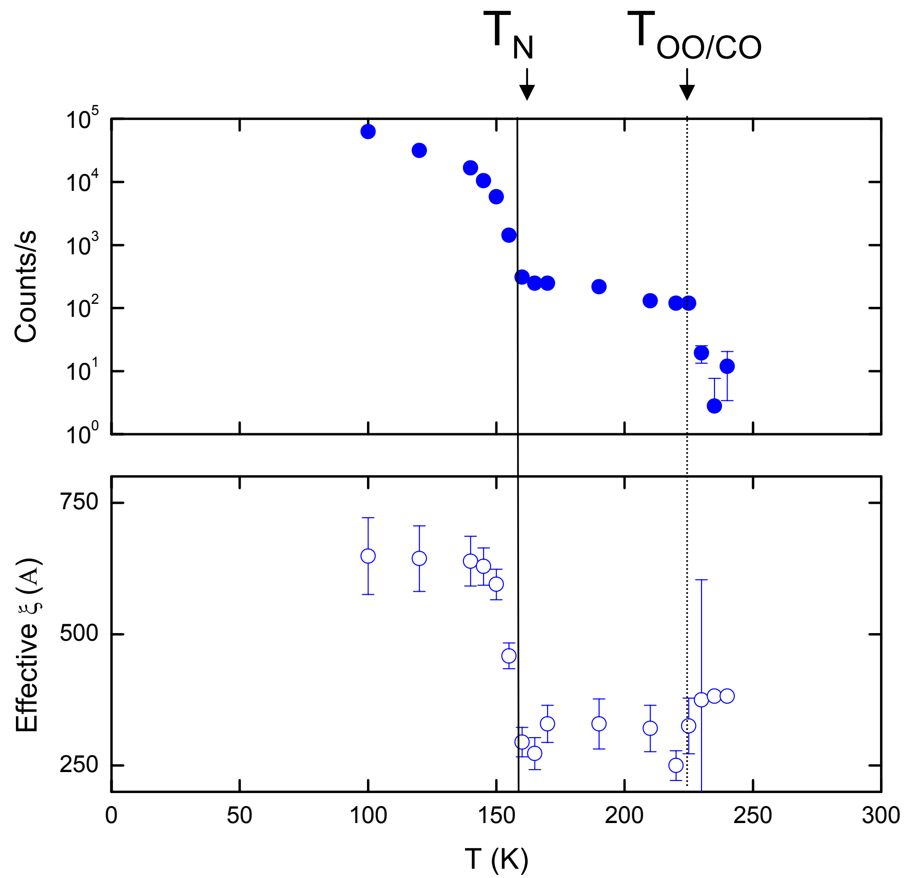
NSLS X1B (P. Abbamonte, L. Venema, G. Sawatzky)

E = 645 eV (L_3 edge)



c out of the diff. plane

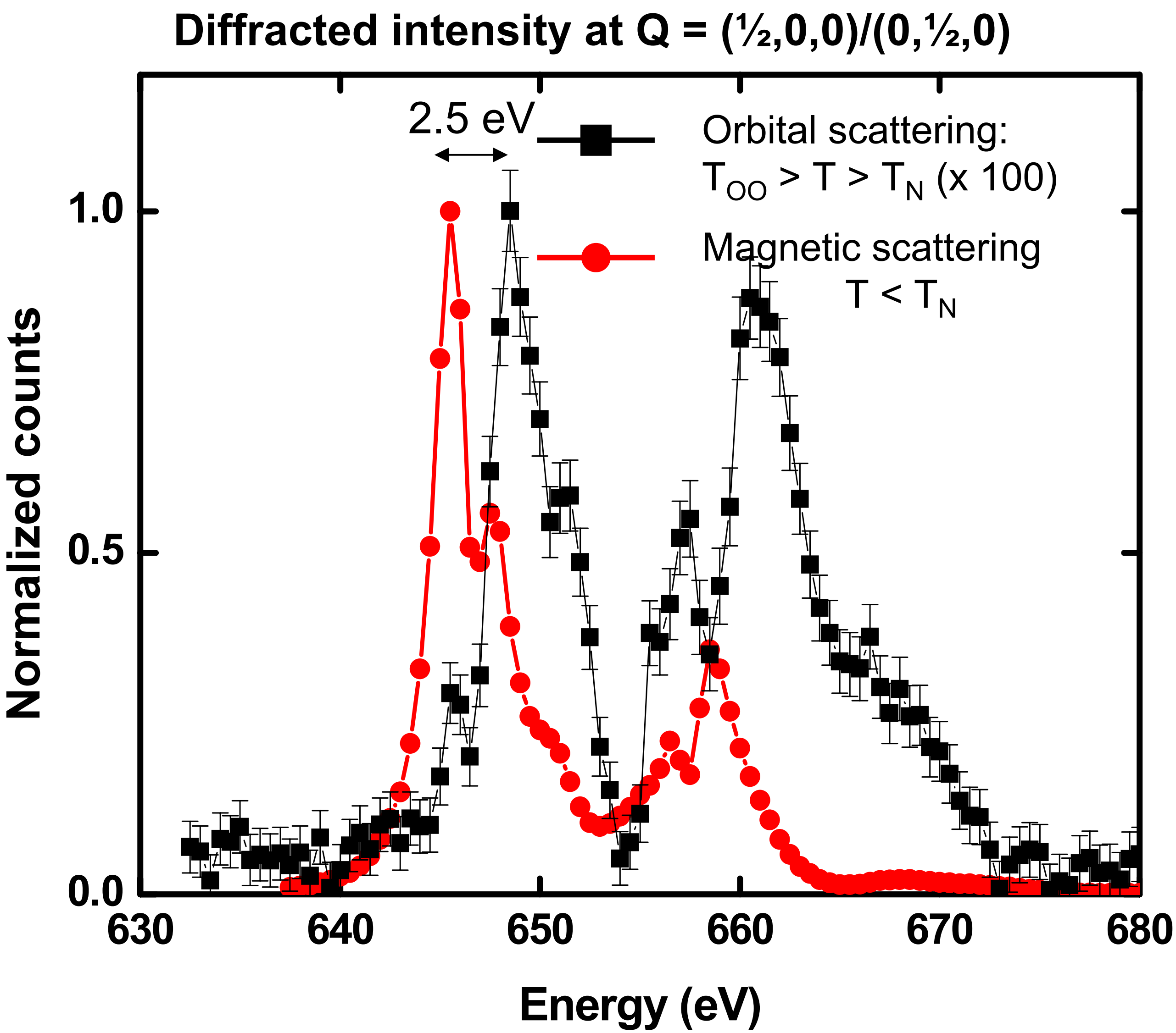
Temperature dependence



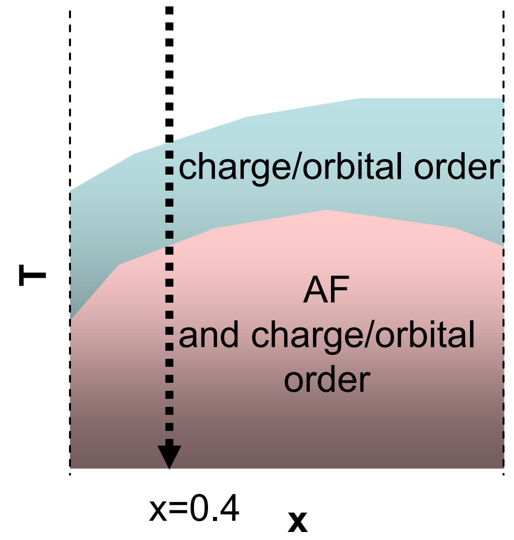
orbital order appears to be correlated over a shorter length scale than magnetic order

K. J. Thomas, J. Hill, S. Grenier, P. Abbamonte, M. v. Veenendaal, G. Sawatzky *et al*. PRL **92**, 237204 (2004).

Magnetic and orbital resonant line shapes



$Pr_{0.6}Ca_{0.4}MnO_3$

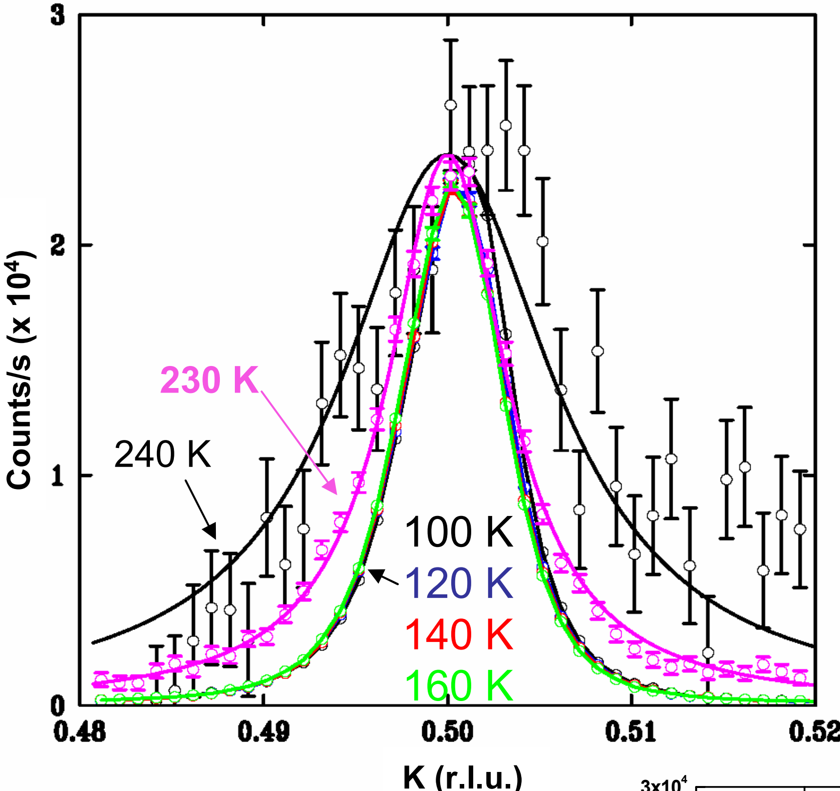


c out of the diff. plane

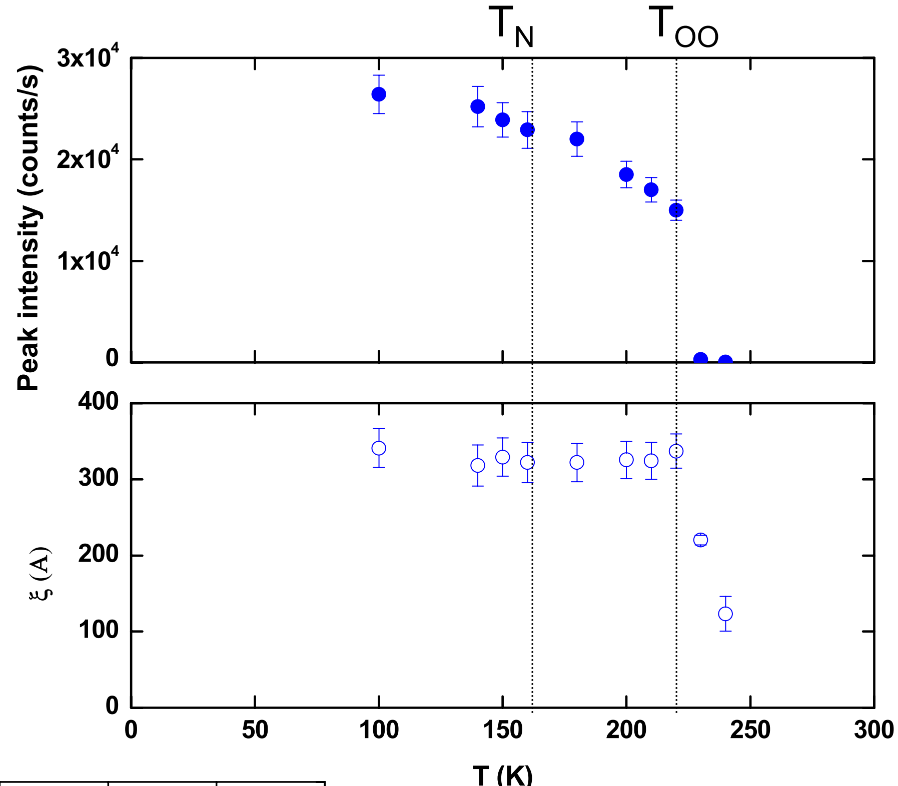
K. J. Thomas *et al*. PRL **92**, 237204 (2004).

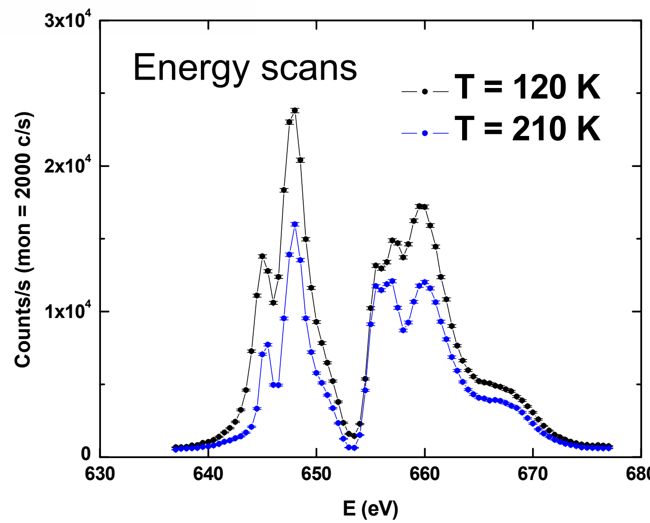
Strong orbital scattering (c in the scattering plane)

Orbital reflection at different T

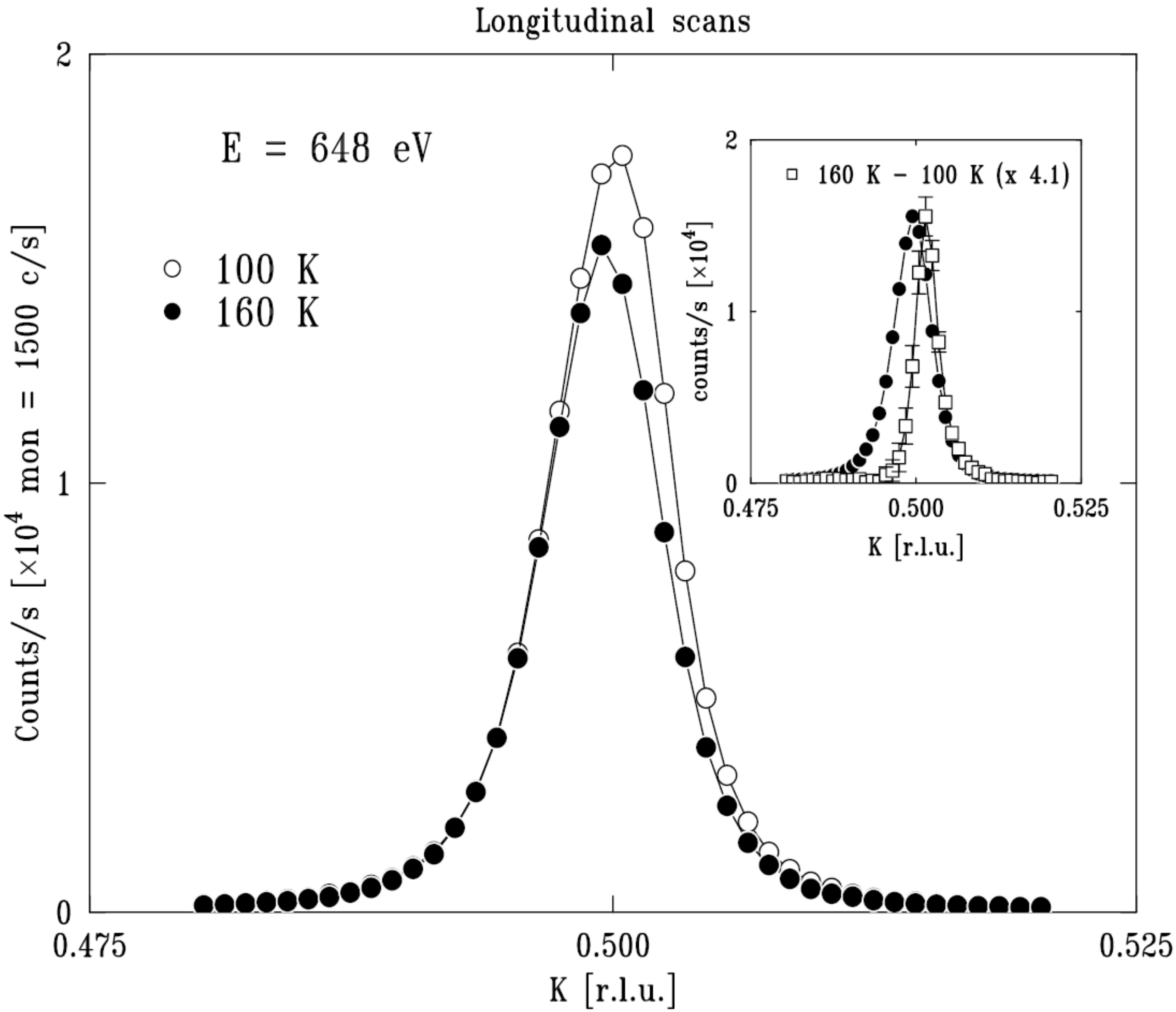


Temperature dependence

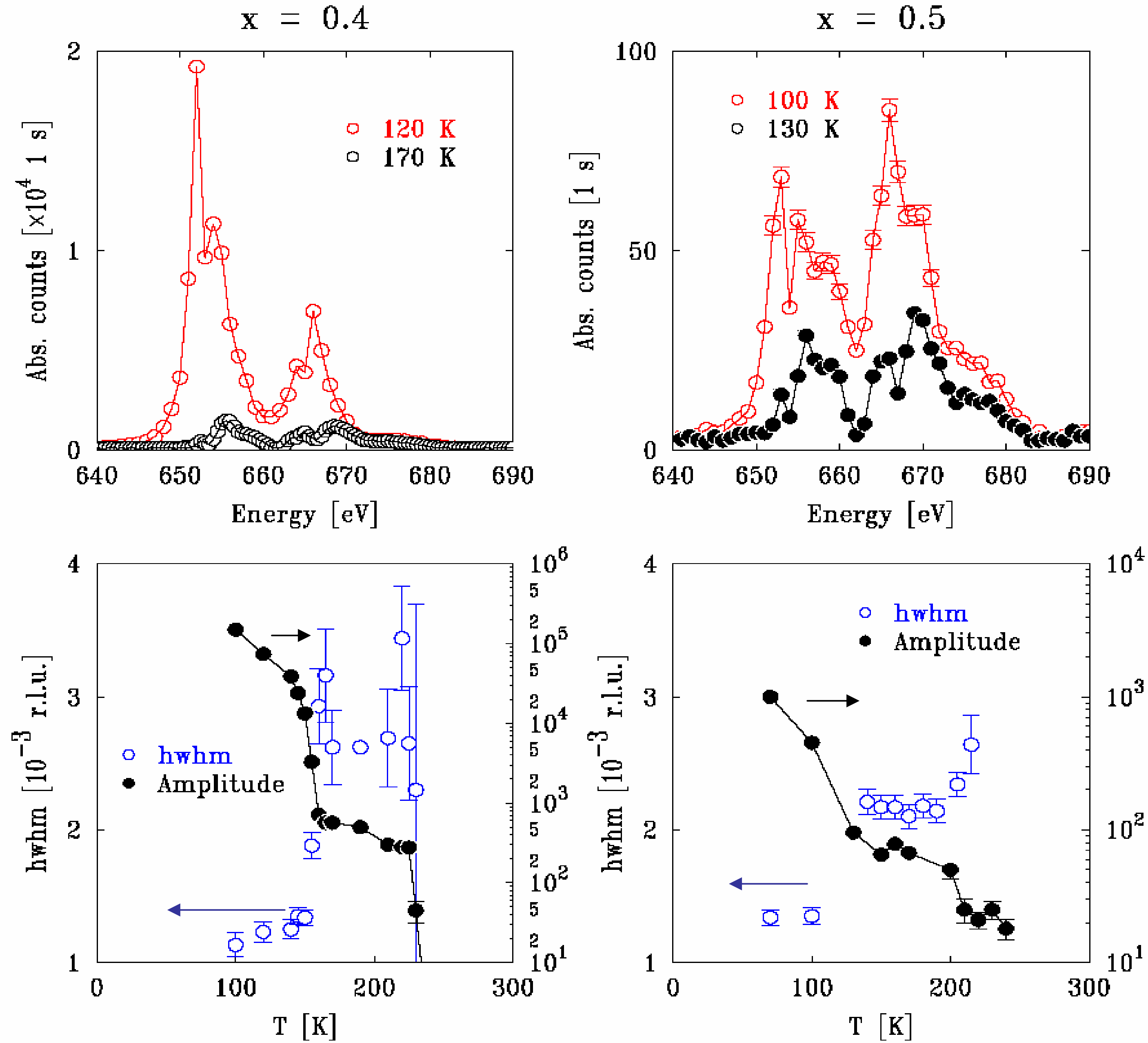




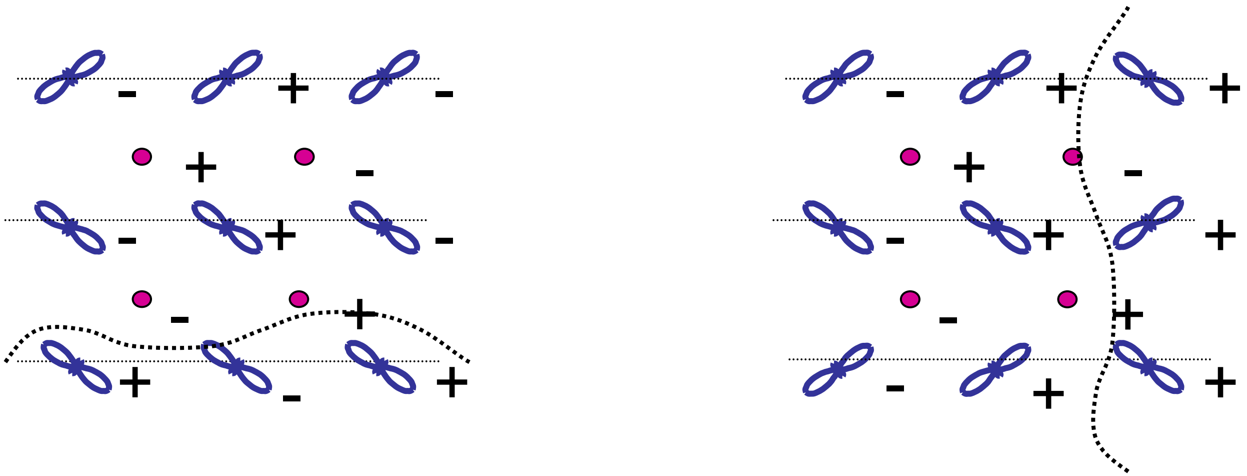
Comparison of magnetic and orbital peak widths: c in the diff. plane



Magnetic and orbital order for different doping: $Pr_{1-x}Ca_xMnO_3$

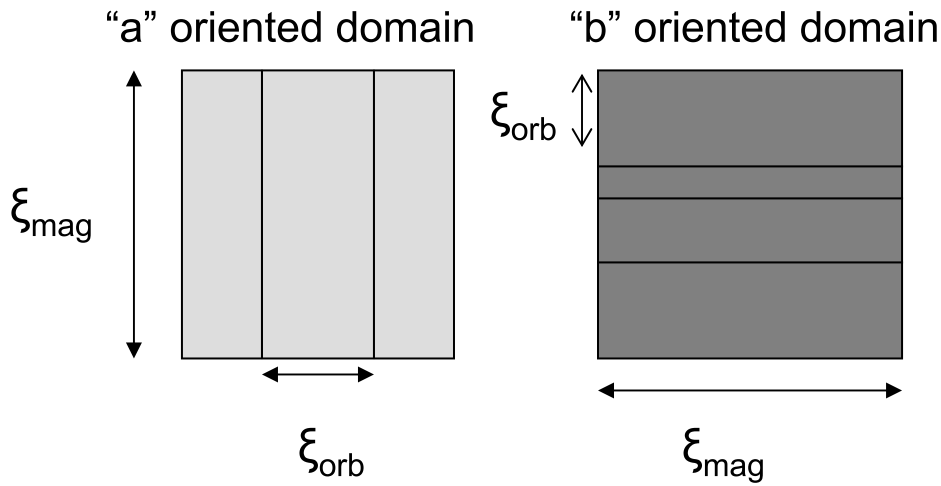


Difference between magnetic and orbital correlation lengths



Difference in correlation lengths

Could the domains be anisotropic?

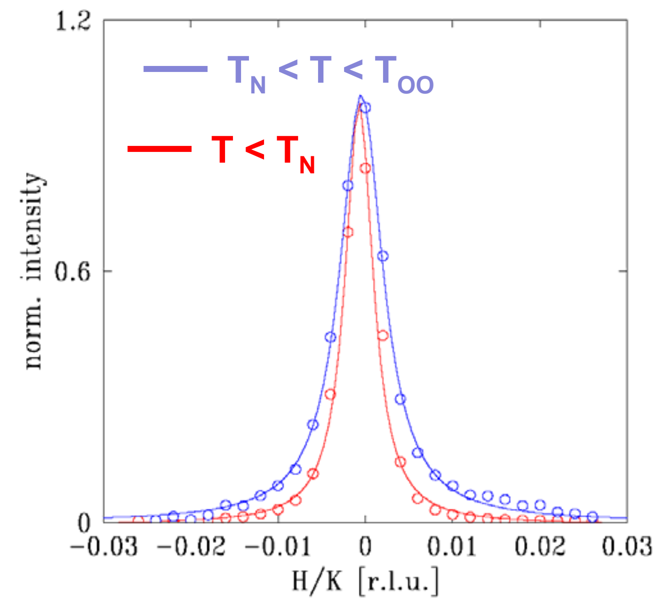


However . . .

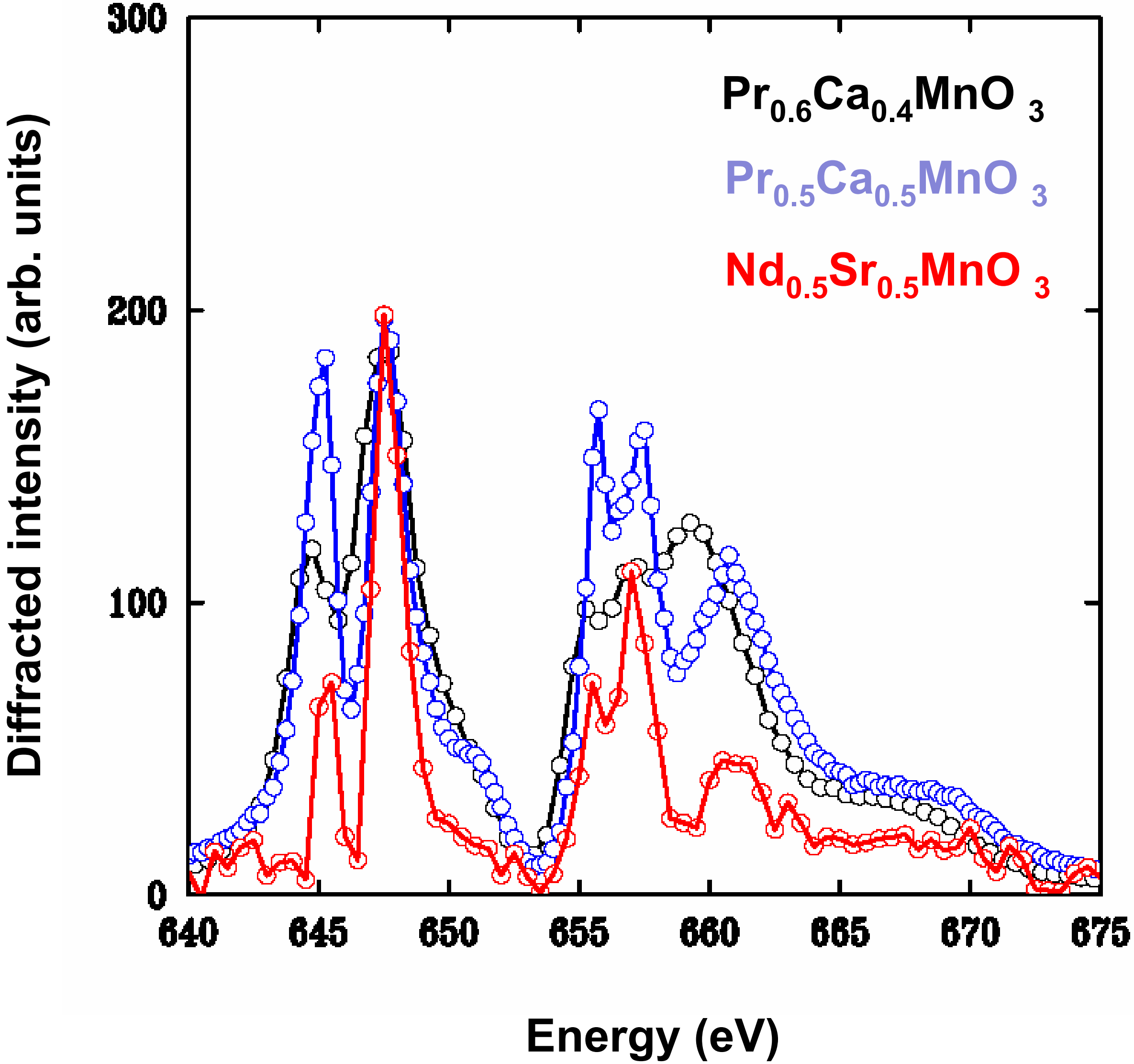
Transverse scans through the orbital
and magnetic
Bragg peaks show a
similar difference in widths

K-edge measurements show
orbital domains are isotropic

Transverse scans



Characteristic orbital diffraction in half-doped manganites



- Similarity of spectra suggest a “thumbprint” on orbital order

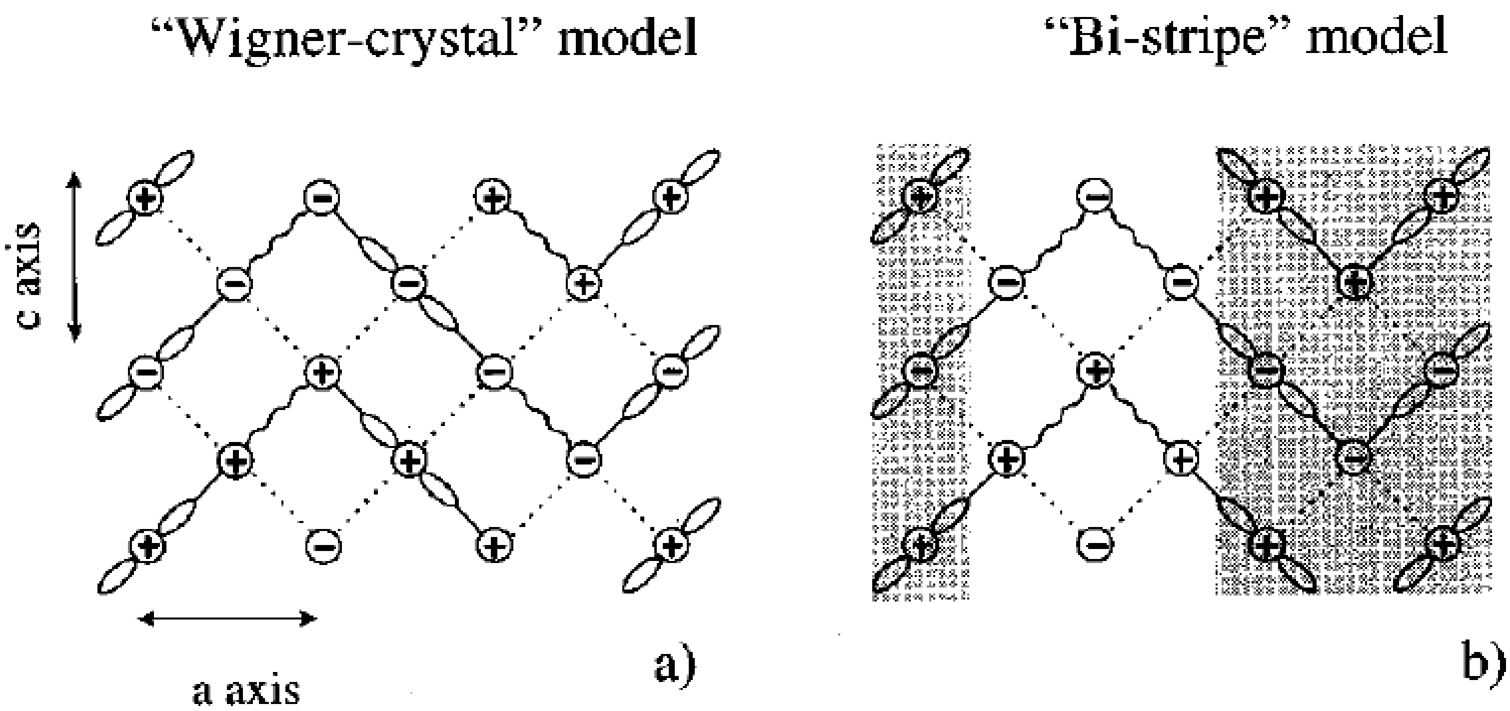
- Improve calculations to isolate features in the spectra (crystal field and hybridization effects)

Probing orbital order at the oxygen K-edge: x > 0.5 manganites

- Electronic models suggest significant hole density on the oxygen sites – can it be measured in a diffraction signal?

O K-edge ~ 540 eV → Only long periodicities can be studied

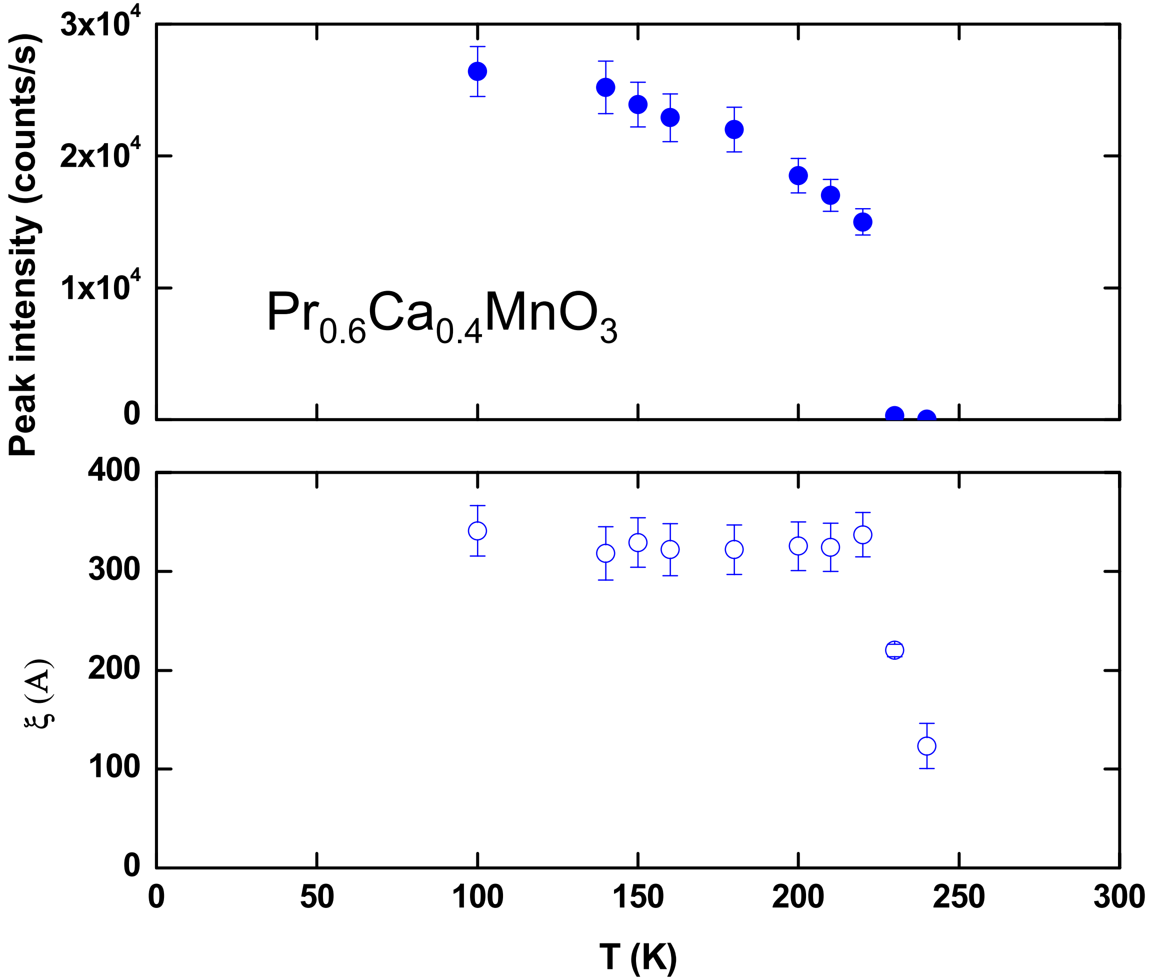
$Bi_{1-x}Ca_xMnO_3$ (x ~ 2/3)



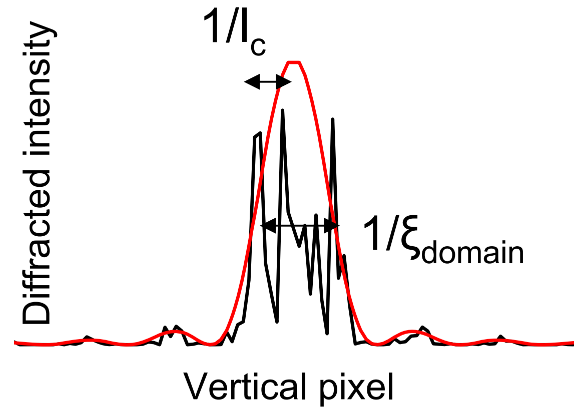
P. G. Radaelli *et al.* PRB 59 14440 (1999)

Mn K-edge studies: S. Grenier and V. Kiryukhin, *unpublished*

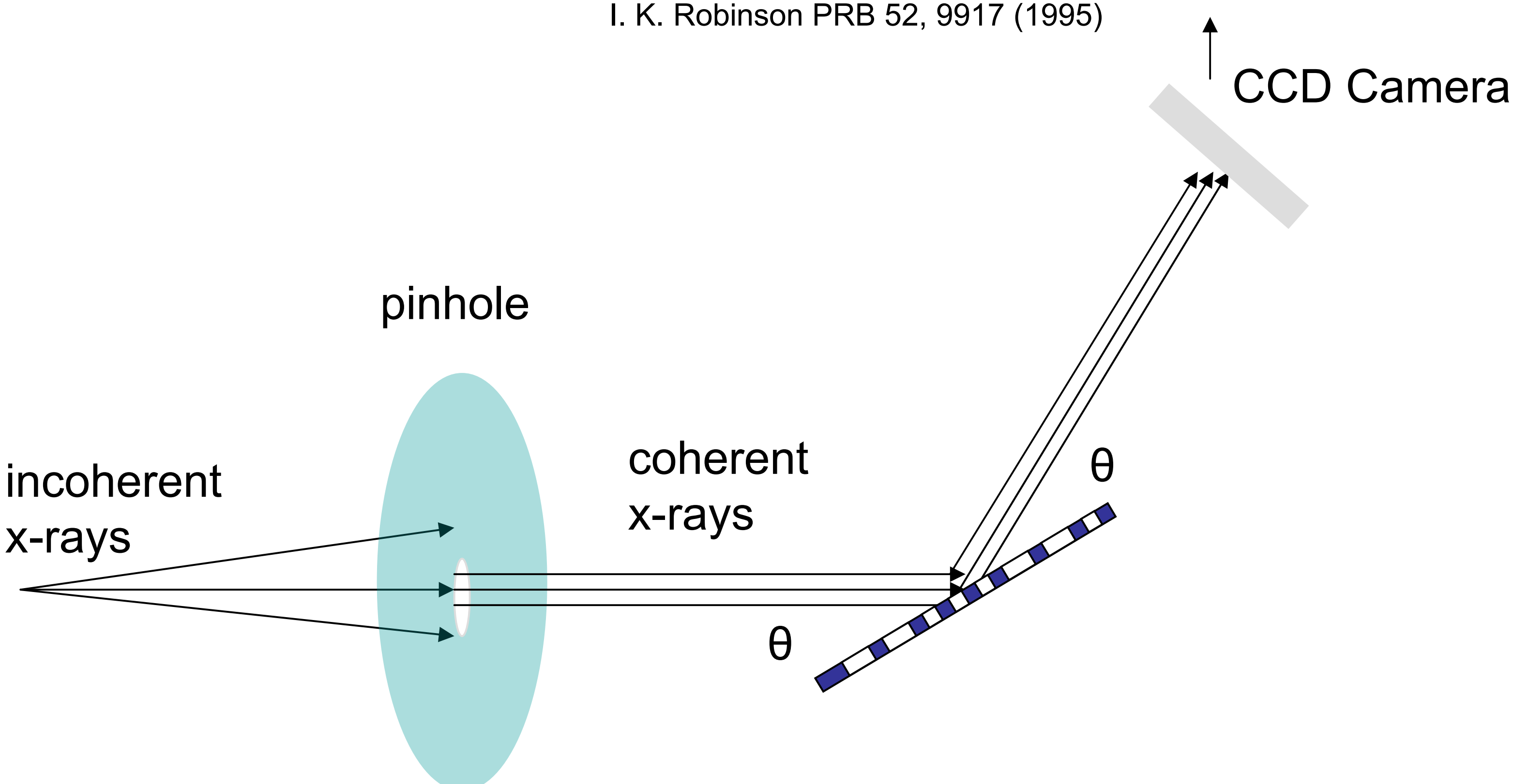
What is the nature of the short-range orbital order?



Probing orbital domains with coherent resonant diffraction

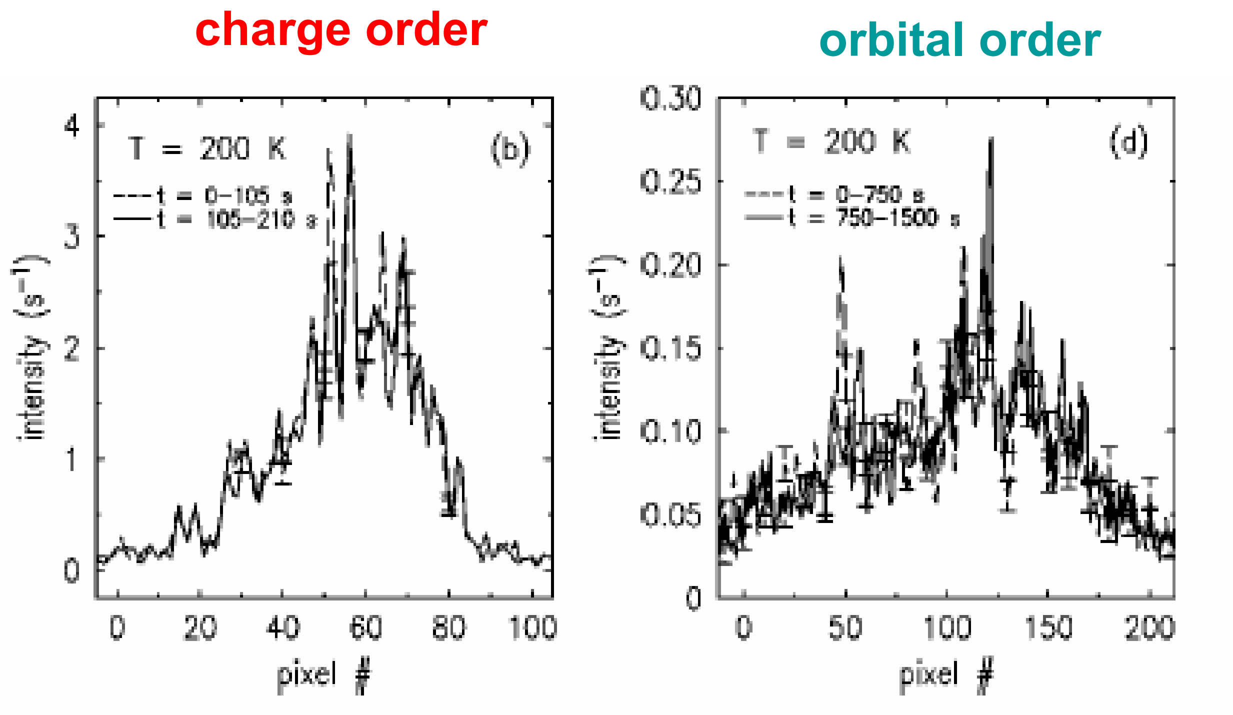


I. K. Robinson PRB 52, 9917 (1995)



Coherent x-ray diffraction of charge and orbital domains at the K-edge

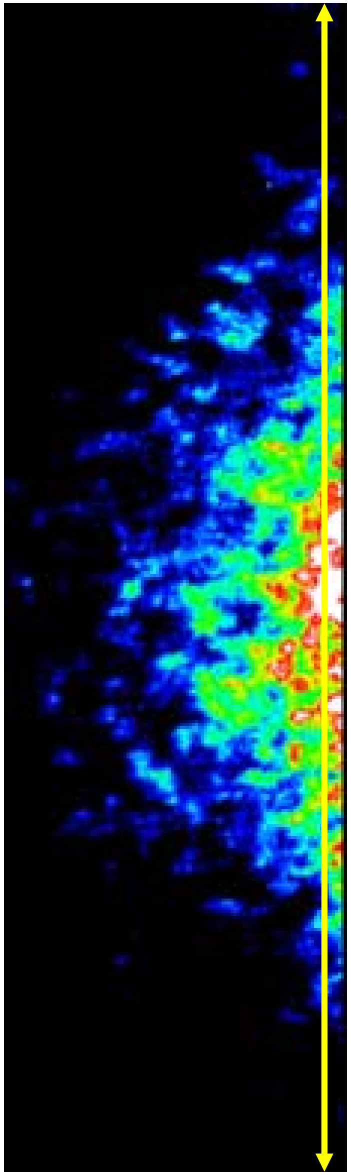
$Pr_{0.6}Ca_{0.4}MnO_3$



C. S. Nelson *et al* PRB 66 134412 (2002)

ESRF ID20

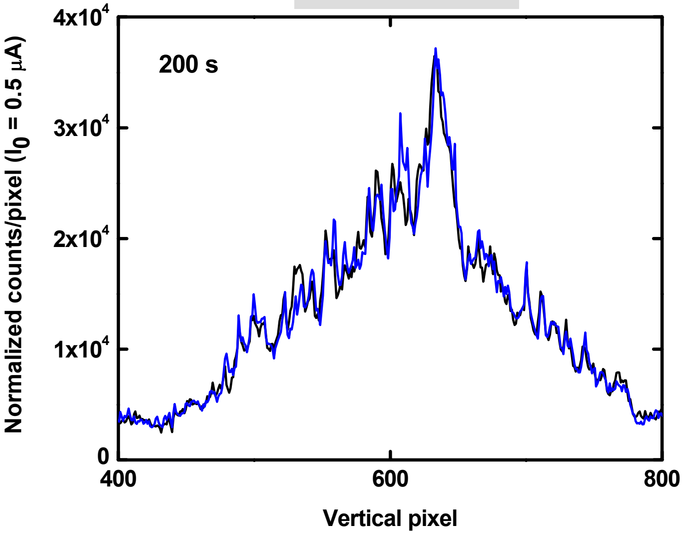
Coherent Resonant Diffraction of Orbital Domains with Soft X-rays



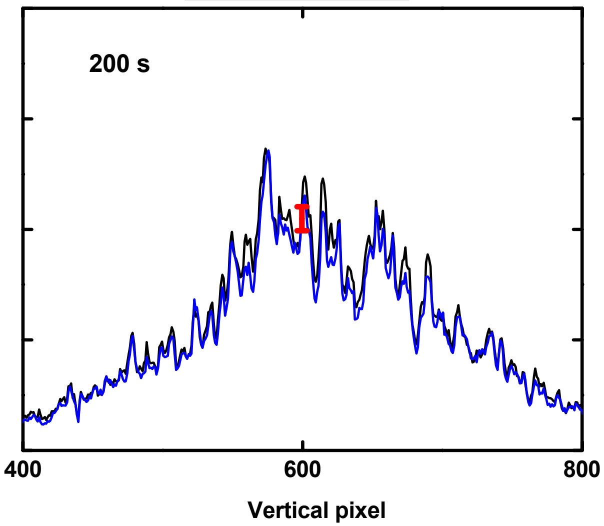
$Pr_{0.5}Ca_{0.5}MnO_3$

Vertical slices through Q_{OO} = (0 ½ 0)

T = 160 K



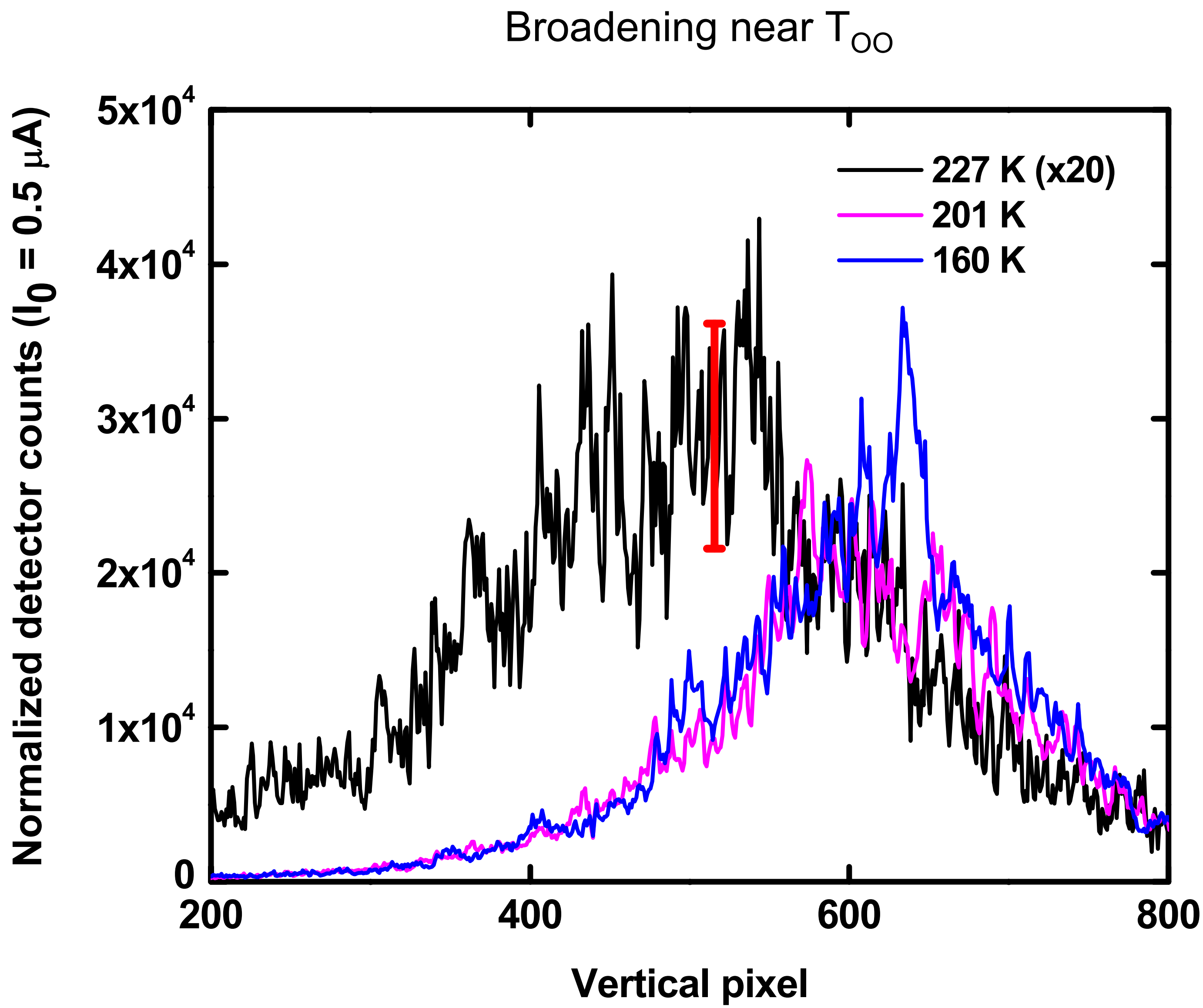
T = 201 K



Orbital speckle appears static to within 20 K of T_{OO}

Advanced Light Source Beamline 12

K. Chesnel and M. Pfeiffer (ALS), J. Thomas and J. Hill (BNL),
J. Turner and Steve Kevan (Univ. of Oregon)


Broadening near T_{OO}
Normalized detector counts (I_0 = 0.5 μA)
Vertical pixel
227 K (x20)
201 K
160 K
0
1x10^4
2x10^4
3x10^4
4x10^4
5x10^4
200
400
600
800

Future experiments

Comparison of magnetic and orbital speckle patterns

Evolution of dynamics as $T \longrightarrow T_c$

Thermal cycling near Tc – are the orbital domains pinned?

Summary

Resonant orbital and spin diffraction in half-doped manganites

- Direct comparison of magnetic and orbital correlations

  Difference between ξ_{orb} and ξ_{mag} suggests magnetic correlations not completely determined by orbital order

- L-edge diffraction in manganites combines spectroscopy with sensitivity to correlations, providing detailed information about the 3*d* electrons and a potential test of ground state models

On-going and future projects

- Coherent diffraction
- Orbital/spin correlations away from half-doping (Mn and O edges)

Collaborators

John Hill *BNL Physics*
Stephane Grenier *BNL Physics*

Wei Ku *BNL Physics*
Dmitri Volja *BNL Physics/SUNY Stony Brook*

Peter Abbamonte *BNL NSLS*
Andrivo Rusydi *BNL NSLS*

Scott Coburn *BNL Physics*
Bill Schoenig *BNL Physics*

V. Kiryukhin *Rutgers University*
C. -W. Cheong *Rutgers University*

Karine Chesnel *ALS Berkeley Lab*
Mark Pfeiffer *ALS Berkeley Lab*
Steve Kevan *The University of Oregon*
Josh Turner *The University of Oregon*

Y. Tokura *University of Tokyo, Japan*
Y. Tomioka *AIST, Japan*

Des McMorrow *University College, London*

Michel van Veenendaal *N. Illinois Univ./ANL*

George Sawatzky *U.B.C.*

Perspectives

Experimental

• Combine high spatial resolution (< 1 μm) with resonant diffraction/microscopy
  - Overcome domain effects
  - Nanopatterning of transition metal oxides

• Spectrometer design
  - High level of reproducibility
  - Develop strategies for sample and spectrometer alignment (permits grazing incidence surface diffraction)
  - Controllable resolution in Q-space

Theoretical

• Connect the resonant line shapes with meaningful quantities (hybridization, electronic structure)